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January 19, 2004

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(1 soft copy to Dr. Ullal; 1 hard copy to Ms. C. Lopez)

Re: Fourth Monthly Report on Pulsed Light Annealing #NDJ-2-30630-11 Mod 6

Dear Harin,

This letter comprises the fourth monthly technical status report for "CIGS Film Fabrication by Pulsed Light Annealing of Precursor Films", which is a task added as Mod 6 to ITN's subcontract #NDJ-2-30630-11, "Plasma-Assisted Coevaporation of S and Se for Wide Band Gap Chalcopyrite Photovoltaics", under the Thin Film Partnership Program. This letter describes work performed during the reporting period of December 15, 2003 through January 14, 2004.

Goals and Approach

The primary objective of this research effort is to demonstrate the production of high-efficiency thin-film CIGS solar cells on polyimide substrates by using high-rate heating from a super-intense pulsed light source. The heating rates to be investigated (millisecond time-scale) are at least two orders of magnitude higher than those reported in previous efforts to use Rapid Thermal Processing (RTP) to convert precursor materials to CIGS films for photovoltaics. Higher heating rates may be advantageous in that (1) thermal degradation of the substrate may be avoided with fast annealing and, (2) diffusion of gallium to the back of the film, which is a major limitation encountered in other CIGS RTP work, may be dramatically reduced. Goals of the present investigation are to determine the viability and challenges of using short (<50 ms) pulses from a super intense light source to:

- Convert sputter-deposited precursor films to chalcopyrite-phase CIGS.
- Improve co-evaporated CIGS electrical properties and thereby allow the use of lower deposition temperatures while retaining device performance.
- Develop a method for CIGS film production that is well suited for production scale-up and capable of producing efficiencies that match those achieved using high-temperature co-evaporation.

An additional goal will be to determine whether high-rate heating can effectively eliminate thru-film and lateral diffusion of elements during conversion of precursor structures to produce CIGS films with high front-side gallium content.

Activities

The fourth reporting period for the Pulsed Light Annealing task coincided with the holiday season. Since ITN Energy Systems, Inc. closes for the week between Christmas and New Years Day, the number of working days during the period was substantially less than for other reporting periods. Nevertheless, progress was made in preparations for the upcoming first round of pulsed light annealing treatments.

Several changes were in the configuration of the sputter deposition chamber to be used for fabricating precursor layer films. In particular, changes were made to address the nonuniformity problems noted in the last report. Theoretical analysis was used to determine a configuration that would give more acceptable thickness nonuniformity in combination with reasonable material utilization. Figure 1 shows that thickness variations measured after the reconfiguration agreed with the theoretical prediction. The new configuration is also predicted to give much better uniformity of the copper-to-indium ratio across the substrate. A second configuration change made to the system was addition of a shutter to block the flux of selenium vapor to the substrate during heat-up and cool-down of the selenium source.

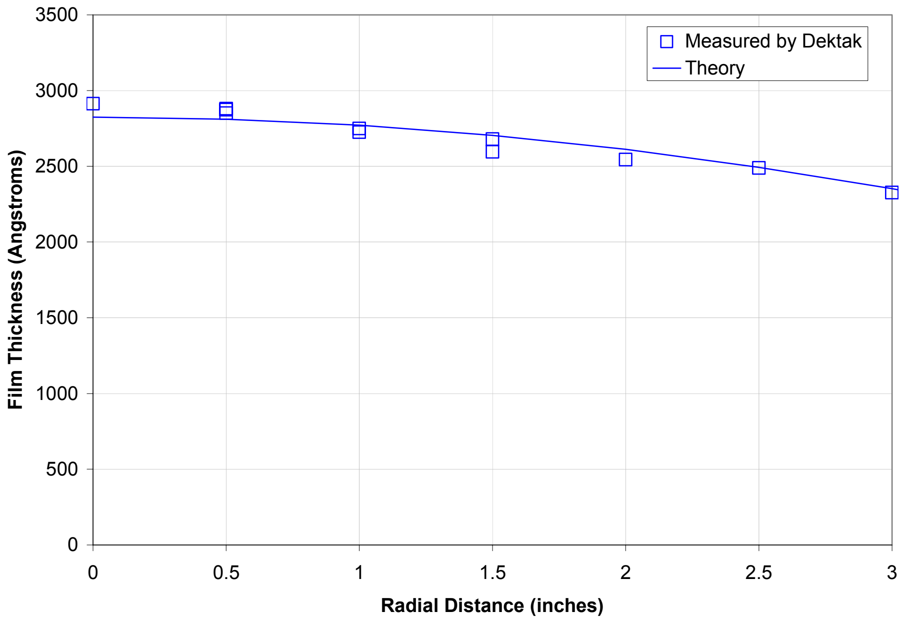

Figure 1: Copper film thicknesses as measured using a Dektak profilometer at various points on the substrate as a function of distance from the substrate's axis of rotation.

Refinements were made to the models for predicting temperature profiles within the CIGS precursor films and substrates. One refinement was incorporating layers to represent the molybdenum and CIGS materials. A second refinement was replacing the ideal square pulse with the actual pulse shape for 20 millisecond pulses. These refinements somewhat increase the fluence (radiant energy density) required to obtain a specified maximum film temperature and decrease temperature gradients within the substrate. Results are shown in Figure 2 through Figure 5 for several cases of interest. In all cases, the pulse fluence values have been adjusted to achieve a maximum film temperature of ~560 °C. Notable features of the results are discussed below.

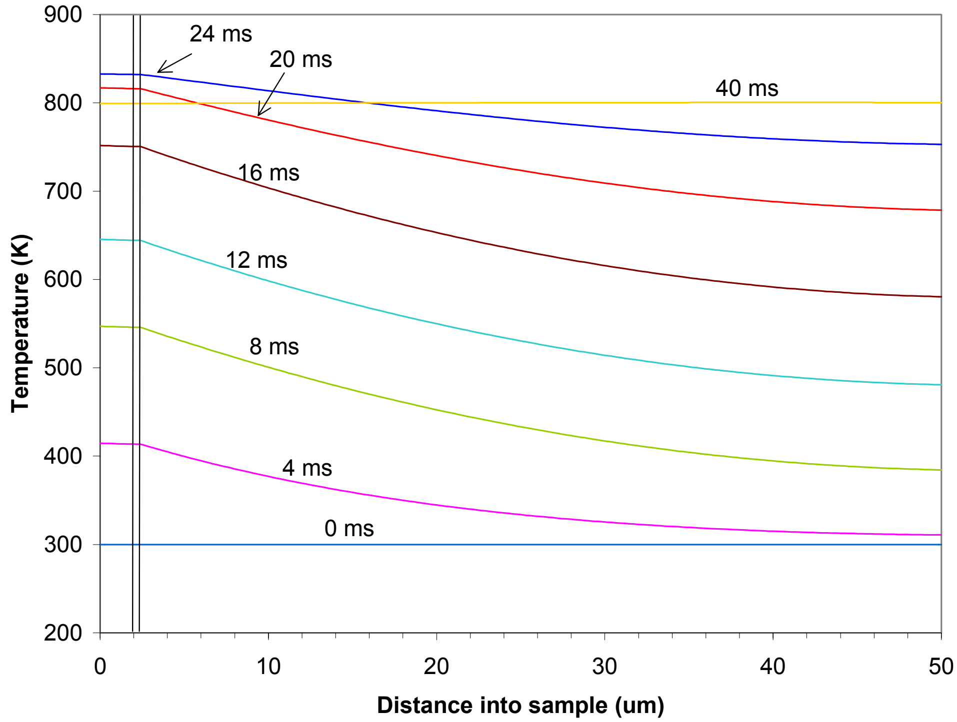

Figure 2: Temperature profiles in a "freestanding" CIGS/Mo-coated polyimide sample during and after a 20-millisecond pulse.

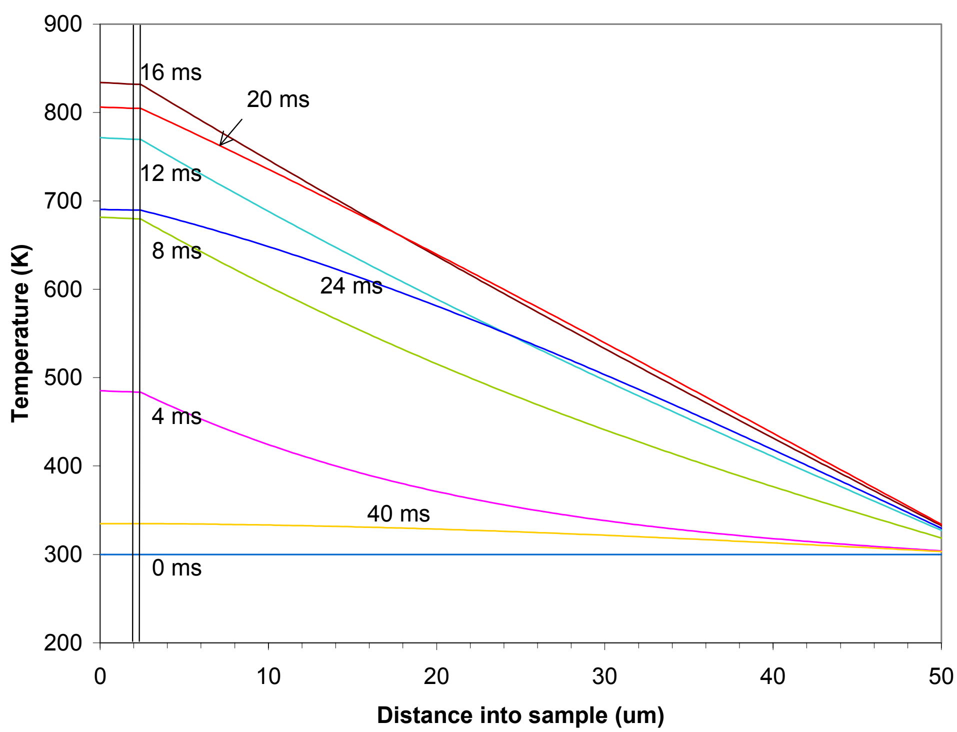

Figure 3: Temperature profiles in a "backed" CIGS/Mo-coated polyimide sample during and after a 20-millisecond pulse.

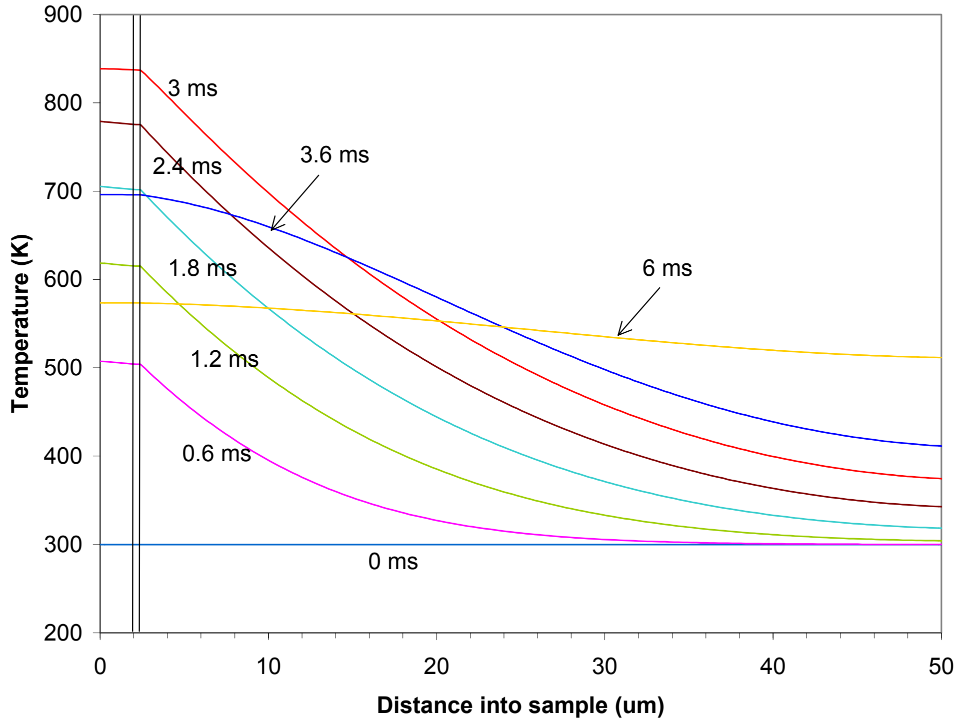

Figure 4: Temperature profiles in a "freestanding" CIGS/Mo-coated polyimide sample during and after a 3-millisecond square pulse.

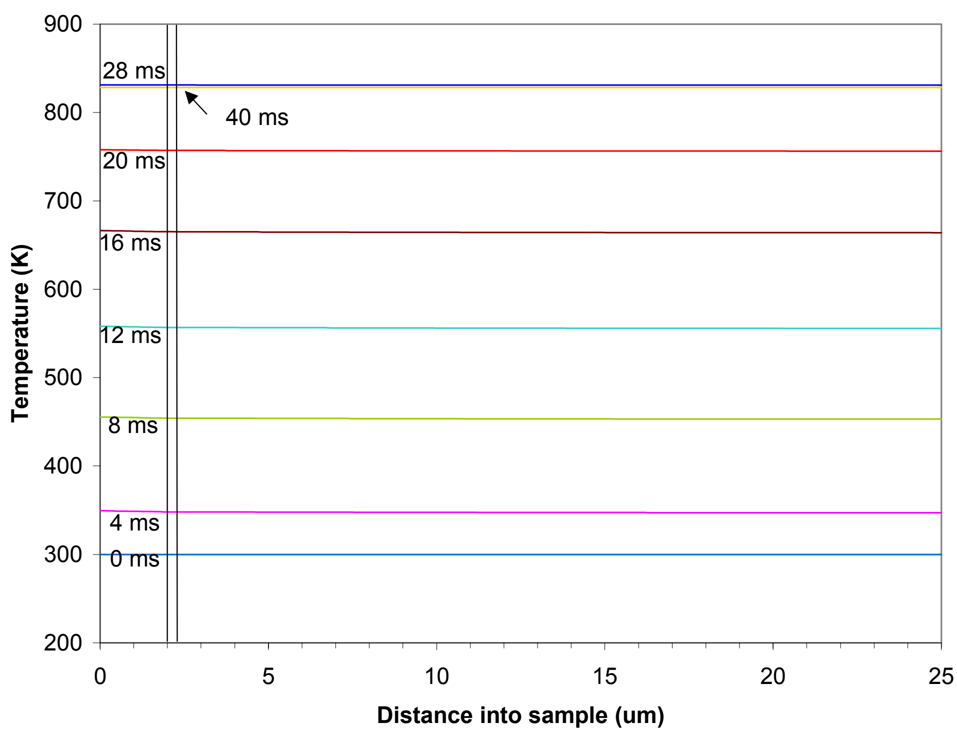

Figure 5: Temperature profiles in a "freestanding" CIGS/Mo-coated stainless steel foil sample during and after a 3-millisecond square pulse.

Figure 2 shows temperature profiles for a “freestanding” polyimide sample subjected to a 20-millisecond pulse. By “freestanding” we mean that the film is held so that neither front nor back surfaces are in contact with another object except on two edges where it is held. Vertical lines on the plot mark the interfaces between the CIGS, molybdenum, and polyimide. The first two microns consist of CIGS precursor materials while the next 0.3 microns consist of molybdenum. The temperature throughout the CIGS and molybdenum layers are relatively uniform as they have much higher thermal conductivity than the polyimide substrate. The CIGS precursor film reaches its maximum temperature 24 milliseconds after the start of the pulse. After the pulse, the heat rapidly redistributes to make a uniform temperature throughout the sample, as shown at 40 milliseconds. The entire polyimide sample reaches a temperature that is only ~30 °C lower than the maximum CIGS precursor film temperature. Whether polyimide can withstand this temperature is a concern.

Figure 3 shows a similar situation except that the sample is held with its backside in intimate thermal contact with a large conductive thermal mass (such as a block of copper). The thermal mass effectively pins the temperature at the back of the sample causing much larger thermal gradients. While the larger stresses associated with these thermal gradients are a concern, the “backed” configuration remains of interest because of the fact that the majority of the polyimide substrate will never be exposed to temperatures exceeding 400 °C, and substrate degradation is therefore less likely.

Figure 4 shows that similar temperature gradients could be achieved in the freestanding configuration if the pulse width is reduced to ~3-milliseconds. The model currently assumes an ideal square pulse because I do not have actual intensity as a function of time for the 3-millisecond pulse. The 3-millisecond pulse in the freestanding configuration may be preferable to the 20-millisecond pulse in the backed configuration due to the likelihood of encountering poor reproducibility with the “backed” configuration as a result of varying degrees of thermal contact between the polyimide and the backing mass.

With a stainless steel foil substrate, there are no concerns with regards to degradation of the substrate at high temperatures as there are with polyimide substrates. Figure 5 shows that there will also be much lower thermal gradients within the stainless steel substrates due to its higher thermal conductivity.

Based on the results described above, a plan has been written for conducting the first round of flash-lamp annealing experiments. Fabrication of parts and samples for these experiments has begun. We are now targeting to complete the first round of flash-lamp annealing procedures in late February.

Best Wishes,

Garth Jensen
Co-Principal Investigator
ITN Energy Systems

Cc: Ms. Carolyn Lopez; NREL contracts and business services